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Supplementary information

Balancing interfacial reactions to achieve long cycle life in high-energy lithium metal batteries

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Supplementary Information

Balancing Interfacial Reactions to Achieve Long Cycle Life in High Energy Lithium Metal Batteries

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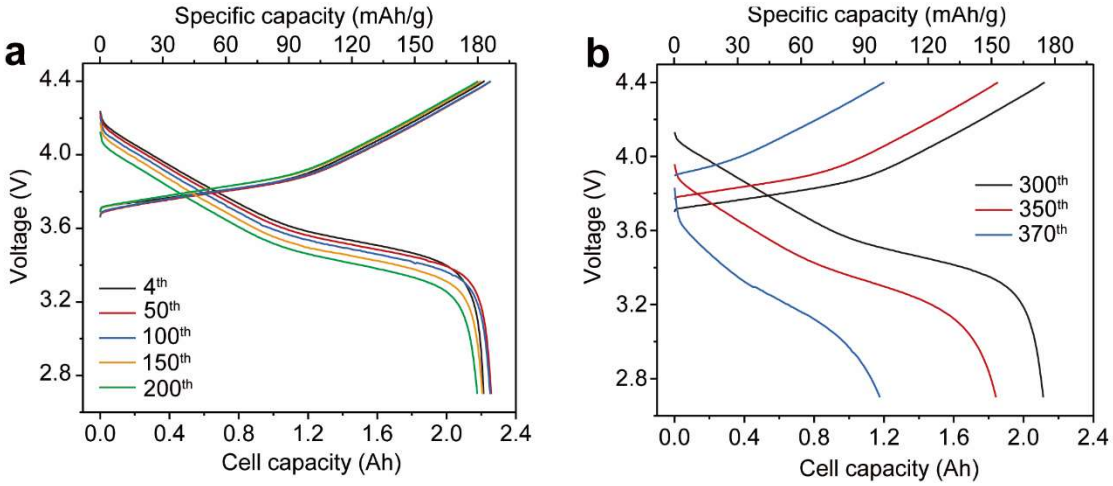
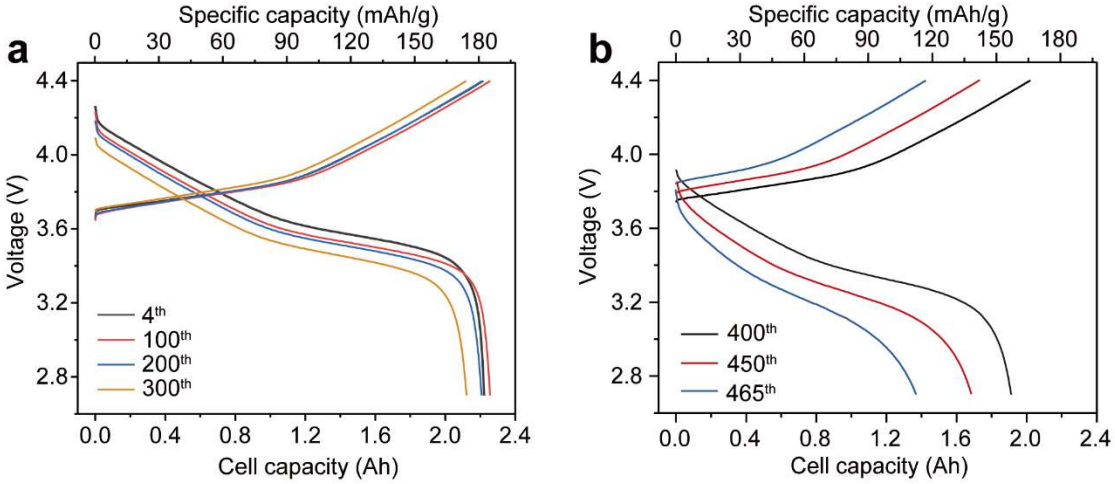
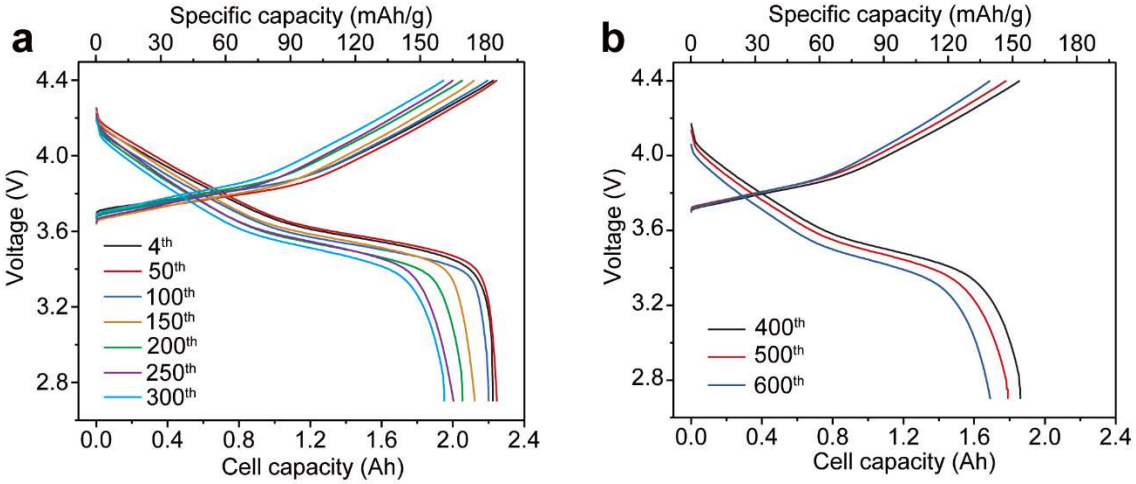
Additional charge/discharge curves of the pouch cells with different Li thicknesses are provided below to clearly show the capacity and voltage change during the early stages and after long cycles. These results clearly show the polarization behavior with a thick Li anode, but the charge/discharge curves for the thin Li anode cell are pretty stable all the way to more than 600 cycles.



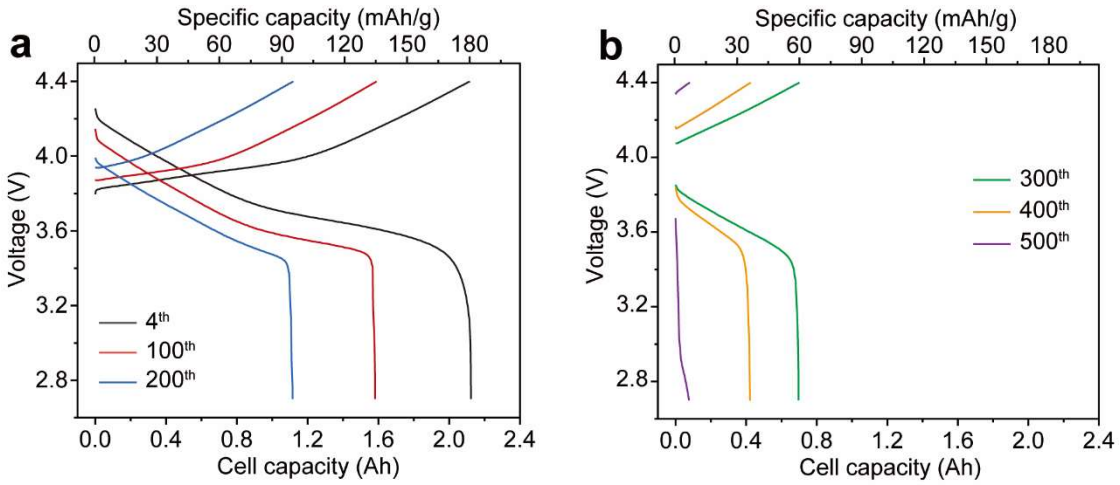
Supplementary Figure 1. The charge/discharge curves of 100 µm Li pouch cell during the initial stable cycling region (**a**) and the fast capacity fading region (**b**).



Supplementary Figure 2. The charge/discharge curves of 50 μm Li pouch cell during the initial stable cycling region (**a**) and the fast capacity fading region (**b**).

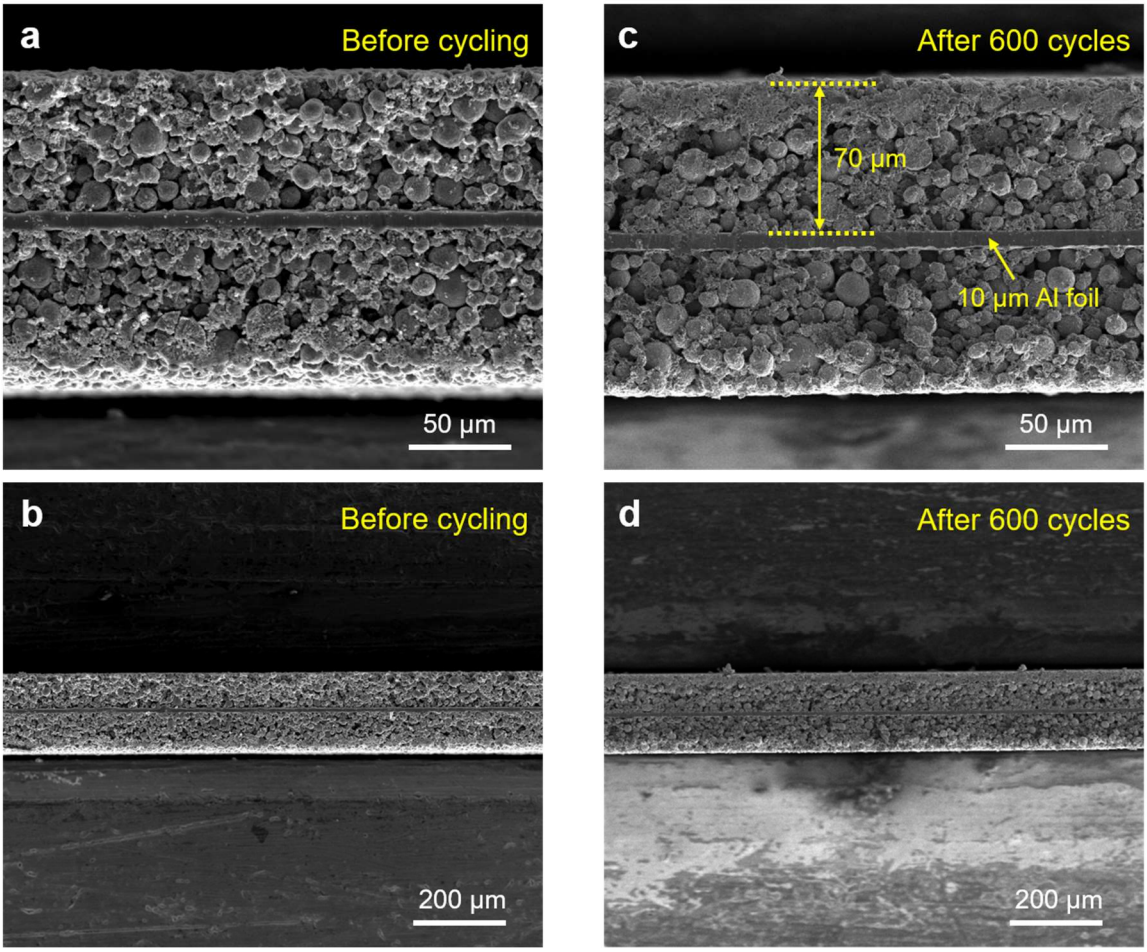


Supplementary Figure 3. The charge/discharge curves of 20 μm thin Li pouch cell, (**a**) from the 4th cycle to 300th cycle, (**b**) from the 400th cycle to 600th cycle.



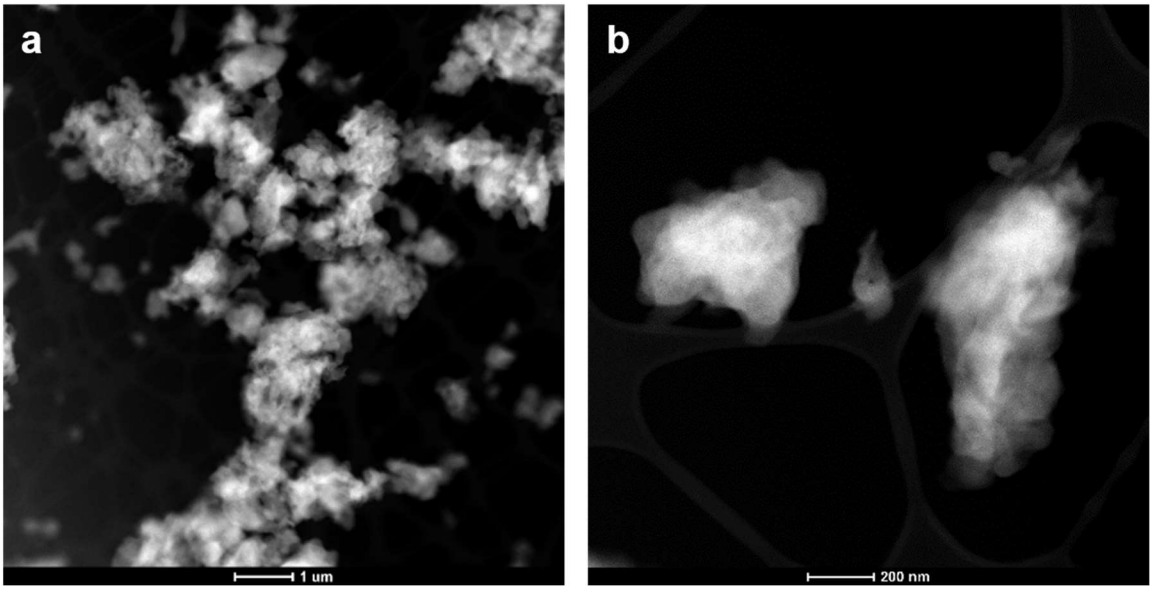
Supplementary Figure 4. The charge/discharge curves of the Li-free-anode pouch cell, (**a**) from the 4th cycle to 100th cycle, 200th cycle, (**b**) from the 300th cycle to 400th, 500th cycle.

Additional cross-sectional SEM images of the cathodes are provided below to demonstrate the stability of the whole cathode architecture. No thickness change or degradation are observed.



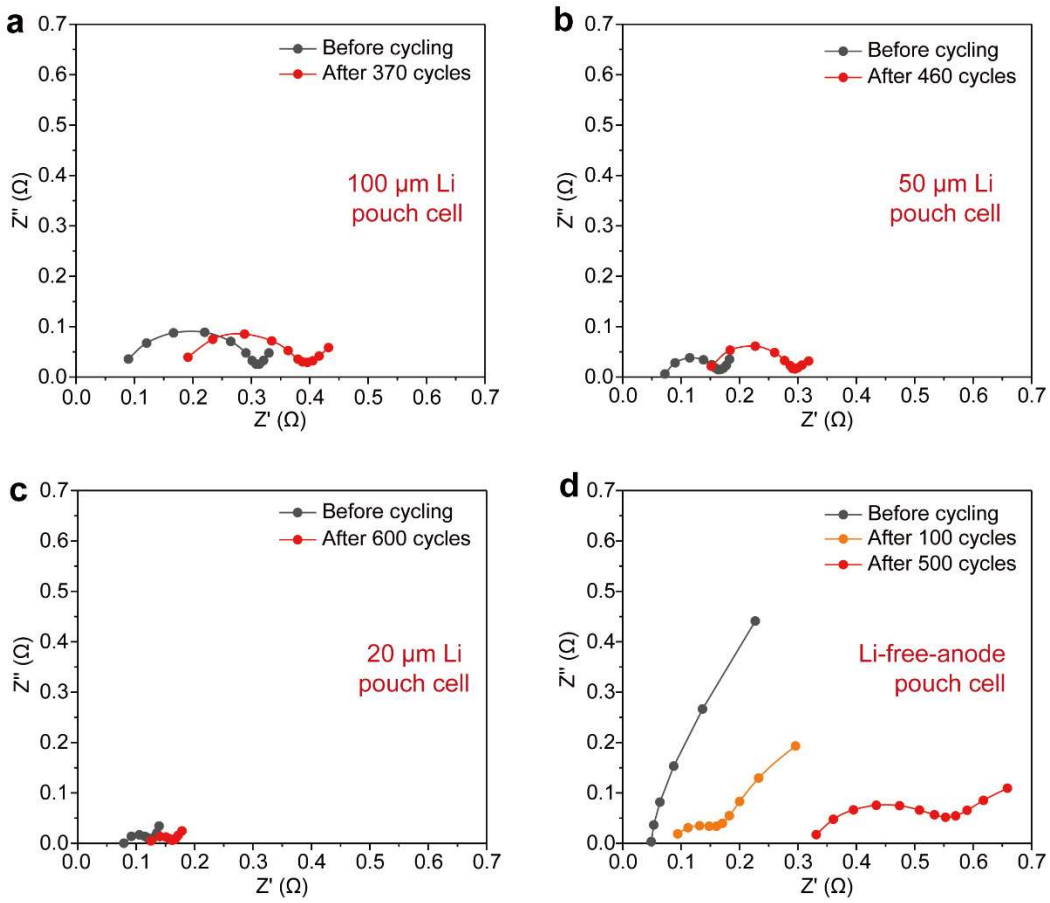
Supplementary Figure 5. The cross-sectional SEM images of one cathode layer (double-sides NMC622 on Al foil) before cycling (**a-b**) and after 600 cycles (**c-d**) in the 20 μm thin-Li pouch cell. The pristine cathode is composed of 96 wt.% NMC622, 2 wt.% conductive carbon and 2 wt.% PVDF binder, the areal capacity on each side is 4.0 mAh/cm^2.

The scanning transmission electron microscope (STEM) images reveal that micron metered sized Li particles are formed after long cycles, and these particles became more and more porous in later stages. These results are consistent with previous reports [25].



Supplementary Figure 6. The STEM images of Li metal particles collected from the anode in 20 μm thin-Li pouch cell after 600 cycles.

Additional impedance measurement results are provided. These results clearly show that the thin Li anode cell has the least impedance at the beginning, and the least impedance increase over long cycles.



Supplementary Figure 7. The impedance measurements of four 350 Wh kg^{-1} pouch cells before and after cycling. a, 100 μm Li pouch cell, **b**, 50 μm Li pouch cell, **c**, 20 μm thin-Li pouch cell, **d**, the Li-free-anode pouch cell. Note that the cathode area is more than 600 cm^2 in each pouch cell, and the pouch cell was initially applied an external uniform pressure of ~172 KPa before cycling. Therefore, the measured cell resistances of pouch cells are much smaller than lab-scale coin cells even with the same chemistry.